Computer simulation of packing of particles with size distributions produced by fragmentation processes

Miguel Angel Martín , Francisco J. Muñoz , Miguel Reyes and F. Javier Taguas

Abstract

Fragmentation schemes inspired in theoretical results and conjectures of Kolmogorov are applied to produce particle size distributions of different nature depending on fragmentation parameters. A 2-D computer simulation method of packing is applied to the resulting distributions and the void fraction is evaluated. The relation between the void fraction and the characteristic parameters of the fragmentation process is studied.

Keywords: fragmentation, granular media, packing, void fraction, particle size distribution.

1. Introduction: Particle size distributions, fragmentation and packing

The study of particulate systems is of great interest in many fields of science and technology. Soil, sediments, powders, granular materials, colloidal and particulate suspensions are examples of systems involving many size particles. For those systems, the statistical description of the particle size distribution (PSD), that is, the mathematical distribution that defines the relative amounts of particles present, sorted according to size, is a central issue. The PSD can be important in understanding soil hydraulic properties, the geological origin of sediments or the physical and chemical properties of granular materials and ceramics, among others.

Several probability distributions have been used as PSD models depending on the nature of the particle system. The log-normal and Weibul distributions have been traditionally used for modelling particle size distributions generated by grinding, milling and crushing operations. The log-hyperbolic distribution was proposed in Bagnold and Barndof-Nielsen (1980) to model PSD of naturally occurring sediments. Dynamical explanations for the occurrence of hyperbolic distribution are also given in that study and factors in the formation of sediments, such as transport by wind or water, are considered. The log-Laplace model was proposed in Fieller et al. (1984) as an alternative for the same scenario.

An important amount of the above mentioned granular media are the result of grinding or fragmentation processes: either natural ones, as in the case of soil and rocks, or artificial ones, as occurs in ceramics or building materials. Some of the models have been postulated on the basis of geometric features observed in such materials. Thus, in the pioneering work of Andreasen and Andersen (1930) the differential equation

$$\frac{dQ}{d(\log r)} = aQ \tag{1}$$

with $a > 0$ a constant, is proposed as a semi-empirical model for the cumulative mass-size distribution function $Q(r)$ of certain granular products with grain size below a given limit. The differential equation is formulated for those products with a grain distribution conformed in such a way that adding a portion of larger grains, the resulting product (grain distribution), is geometrically similar to the previous one, so that, a image of both products seems equal (they have the same "granulography", term coined therein). The model and ability to predict both, the PSD and the void fraction, are tested using data obtained by materials and operations commonly used in ceramics industry.

The log-normal distribution, the most traditional distribution used in particulate media literature, is strongly supported by important theoretical results due to the brilliant mathematician A. N. Kolmogorov. In Kolmogorov (1941) it is mathematically shown that the log-normal distribution for the asymptotic PSD holds when random rules for the fragmentation process are independent of the ratio between the size r of the particle and the size of the particles obtained from it.

In Mandelbrot (1982) and Turcotte (1986) a connection between fractals and fragmentation was made being the start point of a huge number of studies using fractals to model fragment size distributions which have been shown to obey fractal or power law behaviour in many different natural scenarios. It makes justice to say that, surprisingly, the concept of self-similarity or fractality is hidden in the rational basis of the model proposed in Andreasen and Andersen (1930), a half century before Mandelbrot (1982)! In fact, the solution of the differential equation there proposed is a power law. Several models have been proposed to rationalize such empiric evidence. Thus, multiplicative models, together with large deviation theory, have been used to understand these observations suggesting that, the observed power law distributions of fragment sizes should correspond to the superposition of probability density functions

that are log-normal in the "centres" and take the power-law form in the "tail". It would be the result of a natural mixing of simple multiplicative processes that take place along the fragmentation of different particles (Frish and Sornette 1997 and Sornette 2006). However, as was indicated in the above references, there is no accepted theoretical description, although fragmentation processes have a crucial role in the explanation of PSD´s models indeed.

Abstract fragmentation schemes certainly may help in simulating the PSD. However, the result of fragmentation is not easy to predict. An important question was raised in Kolmogorov (1941) about what PSD could be expected when the ratio between the size r of the initial particle and the size of the particles obtained from it has a power law dependence of the size r of the initial particle. This open question remains still without a theoretical answer. Recently fragmentation algorithms inspired in Kolmogorov´s question have been used in Martin et al. (2009) giving results far removed from log-normality showing, on the contrary, great complexity for simulated PSD which are in fact of fractal/multifractal type.

The packing of particles affects the physical properties of the granular system. Properties of particulate materials highly depend on the packing structure. Also, the hydraulic properties (water retention) of granular porous media, as in the case of soil, depend on the bulk density and the geometric scale arrangement of the intergranular space. Even when the particles are modelled by hard spheres, and disregarding the filling material occupying the intergranular space, models of granular media can be useful in predicting different properties of a wide number of natural and engineering systems such as soil, ceramics, porous materials, concentrated suspensions, amorphous materials, alloys or microstructures of simple liquids. Because of that, the study of the random packing of particles has attracted researchers in many areas of science and

technology (see Gray 1968). In particular, the crucial influence of particle size distribution on the random packing structure increases the interest in relating both, either theoretically or by computational methods. Sohn and Moreland (1968) studied the effect of Gaussian and log-normal distributions on packing density using dense random packing of sands, and Roualt and Assouline (1998) used a probabilistic approach to determine the distribution of the volume of the voids in packed spheres once their size distribution was given. Anishchik and Medvedev (1995) used three-dimensional Apollonian packing as a model for dense granular systems investigating the particle size distribution and the fractal nature of packings. The packing of spheres with log-normal distributions by means of computer simulation has been studied in Nolan and Kavanag (1993) and He et al. (1999) used a Monte Carlo simulation for a random model of spherical particles of sizes obeying any given distribution.

The goal of this paper is to explore the connection of the characteristics of fragmentation processes with the packing density of resulting particulate media, and examine this with respect to its important repercussions in many fields of science and technology. First a family of fragmentation algorithms that replicate both, the smooth log-normal and the fractal models, are implemented. Then a 2-D computer simulation method of packing of spheres is applied to the resulting distributions and the void fraction is evaluated. Finally, the relationship between the void fraction and the characteristic parameters of the fragmentation process is studied.

2. Material and methods

2.1 Fragmentation algorithms

Fragmentation algorithms proposed in Martin et al. (2009) provide a wide range of particle size distributions depending of the parameters driving the fragmentation

process. They vary from the log-normal model, when the fragmentation take place with rules that are independent of the size of the fragmented particle, to the high complex fractal/multifractal distributions otherwise.

Let N be a non-negative natural number bigger than one and $\alpha \geq 0$. Each particle of size (volume) r is divided in k smaller particles of size $\frac{r^{1+\alpha}}{k}$, being k a number randomly chosen between 1 and N, with equal probability for all the possible choices. The ratio between the size r of a particle and the size of the particles obtained from it is proportional to r^{α}, certain power of r as suggested Kolmogorov (1941).

The number of particles obtained from a particle of size r is

$$\left\lfloor \frac{r}{\frac{r^{1+\alpha}}{k}} \right\rfloor = \left\lfloor \frac{k}{r^{\alpha}} \right\rfloor \tag{2}$$

and a remaining particle of size

$$r - \left\lfloor \frac{k}{r^{\alpha}} \right\rfloor \cdot \frac{r^{1+\alpha}}{k} \tag{3}$$

where $\lfloor x \rfloor$ denotes the function floor of x, that is the function that returns the greatest integer less than or equal to x.

The algorithm starts by fixing both values of N (integer bigger than one) and α (positive real number such that $0 \leq \alpha \leq 1$), for all the process, and the number of steps p of the fragmentation process. We consider an initial particle of size equal to 1 and in each step we apply, to each particle resulting from the previous step, the following procedure:

1. An integer k is randomly chosen between 1 and N, with equal probability for each one possibility.

2. The considered particle of size r is substituted by $\left\lfloor \frac{k}{r^{\alpha}} \right\rfloor$ particles of size $\frac{r^{1+\alpha}}{k}$ and an additional residual particle of size $r - \left\lfloor \frac{k}{r^{\alpha}} \right\rfloor \cdot \frac{r^{1+\alpha}}{k}$.

2.2 Computer simulation of packing

A Monte Carlo-based method following the model proposed in Vidal et al. (2009) has been considered for the generation of packing processes. This method randomly positions a sequence of particles, ordered by decreasing radius, in a square domain with periodic border and edge length L_0. The center of each particle, with randomly generated coordinates (x, y), will always remain in the domain as long as it does not overlap any previously positioned particle. There exist a maximum number N_{TM} of attempts (of randomly generated (x, y)) of the positioning of each particle within the domain. There also exists a number N_R of the repetition of the positioning of the same sequence of particles within the domain. Once all the particles have been positioned, a new domain will be defined with the length of the smaller edge L_1, where L_1 is defined as of $L_1 = \delta \cdot L_0$, with $0 < \delta < 1$ being a reduction factor of the domain. This positioning process will continue repeating in the new domain L_i, so that $L_i = \delta \cdot L_{i-1}$, until stage n at which a new particle will exist whose number of positioning attempts reaches N_{TM}. At this point the result of the packing simulation of the repetition of the completed N_R will be that obtained at stage $n-1$ where all the particles have been positioned within the domain of size L_{n-1}. The void fraction, V, resulting from that packing will be

$$V = 1 - \frac{S_T}{L_{n-1}^2} \tag{4}$$

where S_T is the total surface area of all the particles of the sequence.

For the execution of the packing simulations, the initial size of the domain, L_0, has been obtained as a function of the total surface area of the particles by means of the

equation $L_0 = (S_T \cdot 2{,}5)^{1/2}$ which produces an initial void fraction $V = 60\%$. The reduction factor δ has been fixed at 0.999 and the maximum number of iterations at 10^6. Each particle sequence repeats $N_R = 10$ times and the value of the ending void fraction V of the simulation for that distribution will be the lowest of these repetitions.

3. Results and discussion

Data coming from Martín et al. (2009) have been used to simulate the packing algorithm. A total number of 47 simulated distributions corresponding to the value $\alpha = 0$ and $N = 2, 3, 4, \ldots, 9$ have been selected. Figure 1 shows the influence of the number of the iterations (in the packing algorithm) on the void fractions. It can be observed that the void fraction has been stabilized for 10^6 iterations, therefore the number of iterations of each packing simulation in this work is 10^6.

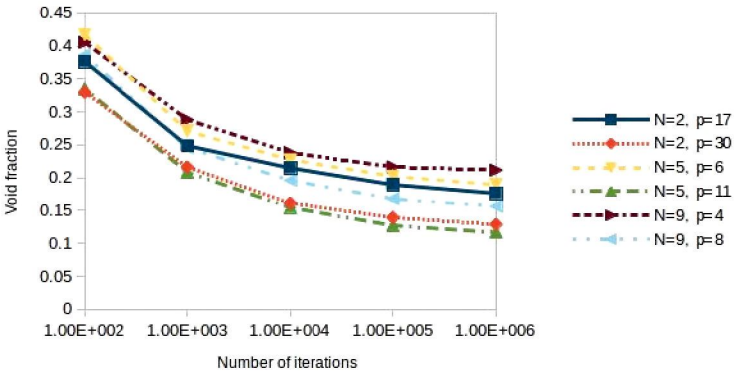


Figure 1: Influence of the number of iterations on the void fraction for some simulations. This behaviour is similar for all values of N and p used.

The log-normality was tested with a Kolmogorov-Smirnov goodness-of-fit test at a confidence level of 95%. In the case $\alpha = 0$ the theoretical mean value and variance may be determined by algorithm parameters and they are given by the formulas

$$E = \frac{-2\sum_{i=1}^{N} i \cdot \ln i}{N(N+1)} \cdot p \qquad \sigma^2 = \left(\frac{2\sum_{i=1}^{N} i \cdot (\ln i)^2}{N(N+1)} - \frac{E^2}{p^2} \right) \cdot p \qquad (5)$$

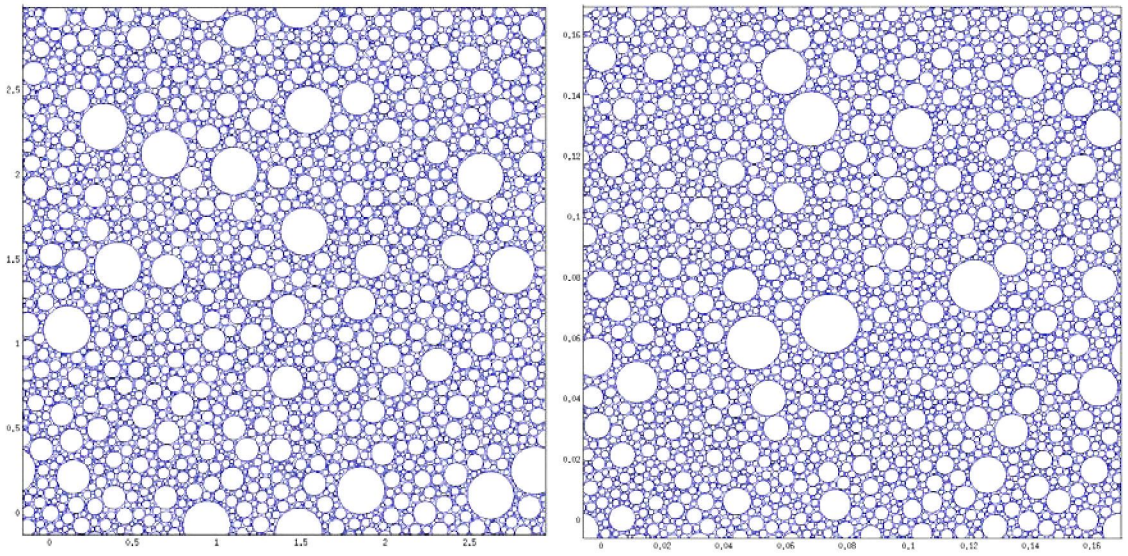
where p is the number of steps of the algorithm and $\ln x$ is the natural logarithm function. Table 1 shows a brief summary of the selected distributions.

N	p	Min	Max	Average	Standard deviation
2	17	3,12E-03	1,41E-01	1,71E-02	1,47E-02
	30	4,87E-05	2,49E-02	9,39E-04	1,19E-03
5	6	5,05E-03	1,03E-01	1,19E-02	7,71E-03
	11	1,01E-04	1,12E-02	8,14E-04	7,76E-04
9	4	8,96E-03	1,51E-01	1,72E-02	1,00E-02
	8	2,37E-04	9,73E-03	1,04E-03	7,47E-04

Table 1: Minimum, maximum, average and standard deviation of the radii of the particles distributions of some simulations.

The theoretical variance given by the algorithm and the real variance of the simulations were computed showing a great agreement (mean error between theoretical and real computed data is 0.285 and the standard deviation is 0.155).

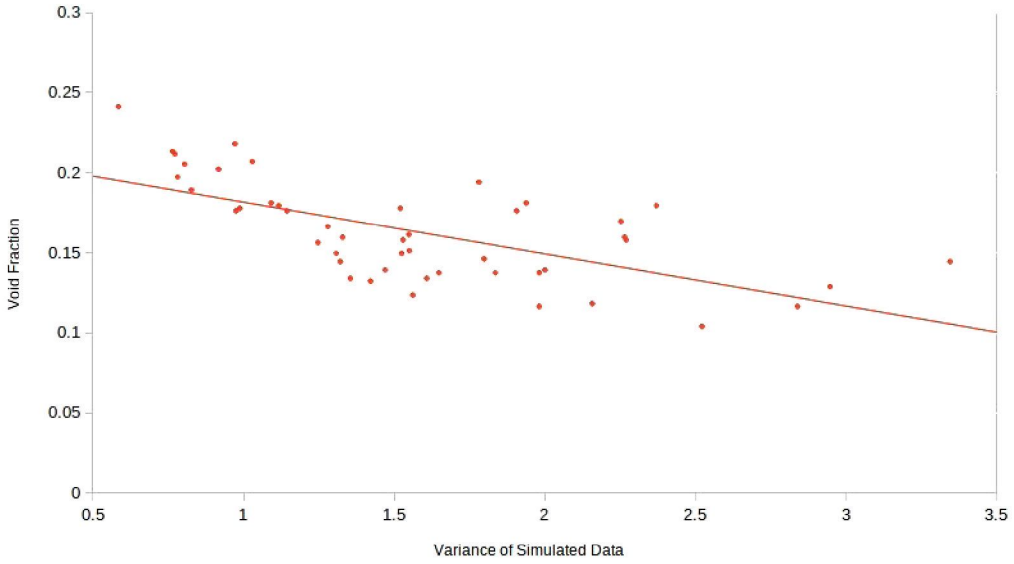
Figures 2 shows some results of the packing algorithm. Figure 2a corresponding to a PSD obtained with $N = 2$ and figure 2b with $N = 9$.



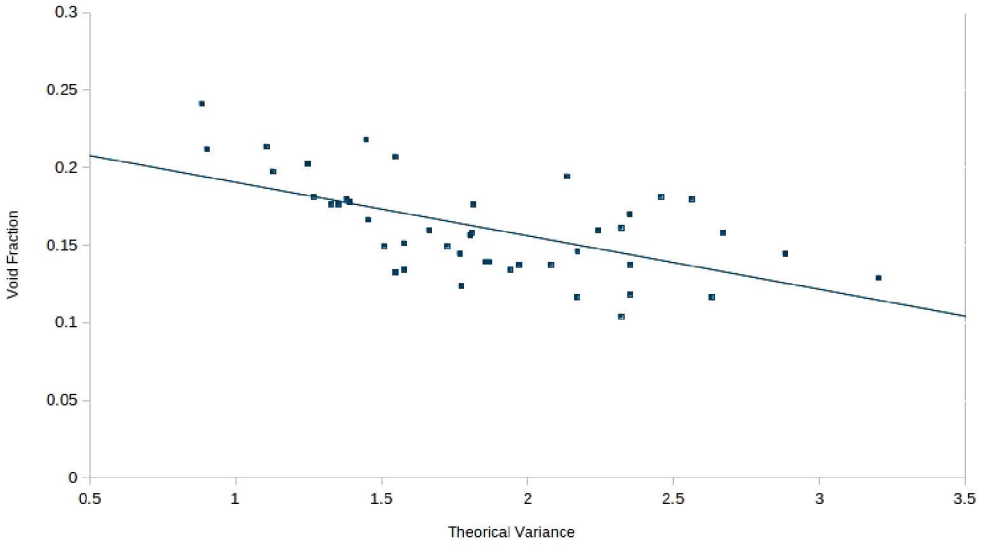
(a) (b)

Figure 2: PSD simulated with packing algorithm with (a) $N = 2$ and (b) $N = 9$

The results respect to the relation between the theoretical and real variance and the void fraction of the simulated packings are shown in figure 3. The determination coefficient R^2 of linear fit are 0.35 and 0.42, respectively.



(a)



(b)

Figure 3: Scatter plots and linear fit of void fraction versus (a) theoretical variance and (b) real variance.

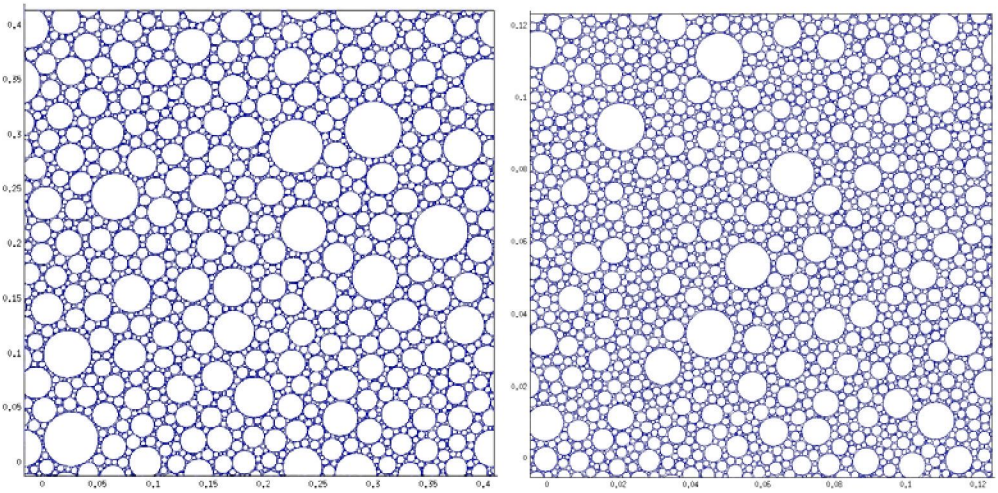
Results clearly indicate there is an inverse correlation between both quantities: the greater is the variance the lesser is the void fraction. This computational result agree with those of Sohn and Moreland (1968) for dense packings of sands.

Also a number of 40 simulated distributions corresponding with different values of α (between 0.1 to 0.5) have been selected to explore how the fragmentation exponent influences the result of packing. Table 2 shows average and standard deviation of void fraction for different values of fragmentation exponent α and parameter N.

N	Fragmentation exponent	Average void fraction	Standard deviation
2	0,1	0,067	0,0038
	0,2	0,058	0,0067
	0,3	0,117	0,0149
	0,5	0,189	0,0030
3	0,1	0,080	0,0029
	0,2	0,081	0,0059
	0,3	0,137	0,0068
	0,5	0,155	0,0031

Table 2: The average and standard deviation of void fraction for different values of α and N

The packing algorithm applied to them. Figure 4 shows some examples of the packing results. Figure 4a corresponding to a PSD obtained with $N = 3$ and $\alpha = 0.1$, and figure 4b with $N = 9$ and $\alpha = 0.5$.

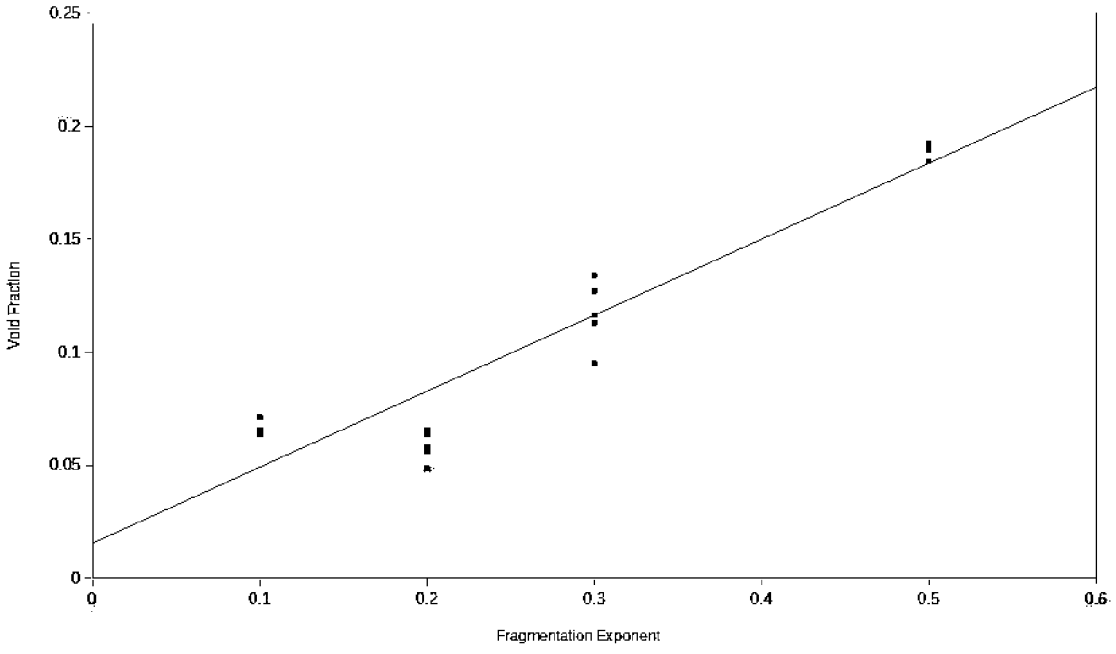


(a) (b)

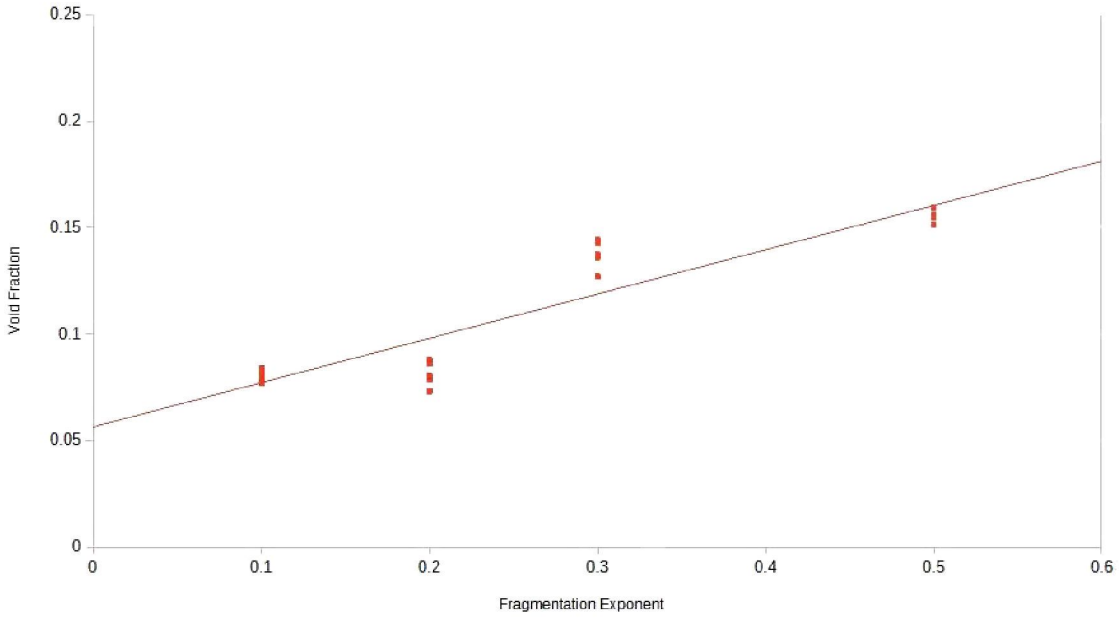
Figure 4: PSD simulated with packing algorithm with
(a) $N = 3$, $\alpha = 0.1$ and (b) $N = 9$, $\alpha = 0.5$.

Figure 5 shows the relation between the value of α and the value of the void fraction for $N = 2$ and $N = 3$. Also the trend lines are shown. The coefficient of determination R^2 of the corresponding fit are 0.893 and 0.835, respectively. Results indicate that the void fraction increase for increasing values of α.

A deeper insight round interesting questions related with the result of the fragmentation process for $\alpha > 0$ (open question posed by Kolmogorov) and the packing arrangement derived are suggested by this result. On one hand a dependence of the scale may be now expected which might explain the fractal nature of the resulting distribution. Secondly, for $\alpha = 0$ the fragmentation of a given particle may give particles of any size with the same probability, small particles in particular, which would facilitate a greater packing density. On the contrary for $\alpha > 0$ the result of fragmentation has not such free rules and depends on the size of the fragmented particle (scale): the size of any particle influences the emergent of new sizes, in particular neighbour sizes which might increase the intergranular space.



(a)



(b)

Figure 5: Plots of void fraction versus fragmentation exponent and trend lines for (a) $N = 2$ and (b) $N = 3$

4. Conclusions

Fragmentation algorithms inspired in results and open problems raised in Kolmogorov (1941) provide a wide range of particle size distributions depending on the parameters driving the fragmentation process. They vary from the log-normal model to the high complex fractal/multifractal distributions. Because of this, they offer an interesting possibility to explore the connection of the characteristics of fragmentation processes with the packing density of resulting particulate media, and to examine this with respect to its repercussions in many scientific fields.

For the log-normal model ($\alpha = 0$), both the theoretical and the real variance of simulations inversely correlate with the void fraction: the greater the variance, the lesser the void fraction.

When $\alpha > 0$ the fragmentation algorithm produces non-smooth complex distributions for which the void fraction increases for increasing values of α. A deeper

insight might indicate that on one hand scale dependence on the result of fragmentation may be expected and, on the other, that such dependence might facilitate a greater intergranular space.

Acknowledgment:

This research work was been funded by Spain´s Plan Nacional de Investigación Científica, Desarrollo e Innovación Tecnológica (I+D+I) under ref. AGL2011–25175.

References

Andreasen, A. and Andersen, J. (1930), *Ueber die Beziehung zwischen Kornabstufung und Zwischenraum in Produkten aus losen Ko¨rnern*, Kolloid-Zeitschrift 50, 217-228.

Anishchik, S.V. and Medvedev, N.N. (1995), *Three-Dimensional Apollonian Packing as a Model for Dense Granular Systems*, Phys. Rev. Lett. 75, 4314-4317

Bagnold, R.A. and Barndorff-Nielsen, O. (1980), *The pattern of natural size distributions*, Sedimentology 27 (2), 199-207.

Caniego, F.J., Martín, M.A. and San José, F. (2001), *Singularity features of pore-size soil distribution: singularity, strength analysis and entropy spectrum*, Fractals 9, 305-316.

Caniego, F.J., Martín, M.A. and San José, F. (2003), *Renyi dimensions of soil pore size distributions*, Geoderma 112, 205-216.

Evertsz, C.J.G. and Mandelbrot, B.B., *Multifractal measures*, In Chaos and fractals (Ed. Peitgen, H-O. et al.) (Springer-Verlag, New York 1992) pp. 921-953.

Feder, J., *Fractals* (Plenum Press, New York 1988).

Fieller, N.R.J, Gilbertson, D.D. and Olbricht, W. (1984), *A new method for environmental analysis of particle size distribution data from shoreline sediments*, Nature 311, 648-651.

Frish, U. and Sornette, D. (1997), *Extreme deviations and applications*, J. Phys. I France 7, 1155-1171.

Gray, W.A., *The packing of solid particles* (Chapman and Hall, London, 1968).

He, D., Ekere, N.N. and Cai, L. (1999), *Computer simulation of random packing of unequal particles*, Phys. Rev. E 60, 7098-

Kinchine A. I., *Mathematuical Foundation of Information Theory* (Dover, New York 1957).

Kolmogorov, A.N. (1941), *On the logarithmic normal distribution of particle sizes under grinding*. Dokl. Akad. Nauk SSSR 31, 99-101.

Mandelbrot, B.B., *The fractal geometry of nature* (W.H. Freeman, New York 1982).

Martín, M.A.and Rey, J.M. (2000), *On the role of Shannon entropy measuring heterogeneity*, Geoderma 98, 1-3.

Martín, M.A., García-Gutiérrez, C. and Reyes, M. (2009), *Modeling multifractal features of soil particle size distributions with Kolmogorov fragmentation algorithms*, Vadose Zone J. 8, 202-208.

Nolan, G.T. and Kavanag, P.E. (1993), *Computer simulation of random packings of spheres with log-normal distributions*, Powder Technol. 76, 309-316.

Peitgen, H.O., Jürgens, H. and Saupe, D., *Chaos and fractals* (Springer-Verlag, New York 1992).

Renyi, A., *Statistical Decision Functions and Random Processes*, In Trans. 2nd Prague Conf. on Information Theory, pp. 545-556.

Roualt, Y. and Assouline, S. (1998), *A probabilistic approach towards modeling the relationships between particle and pore size distributions: the multicomponent packed sphere case*, Powder Technol. 96, 33-41.

Shannon, C.E. (1948), *A mathematical theory of communication*, I. Bell Syst. Tech. J. 27, 379-423.

Sohn, H.Y. and Moreland, C. (1968), *The effect of particle size distribution on packing density*, Can. J. Chem. Eng. 46, 162-167.

Sornette D., *Critical phenomena in Natural Sciences* (Springer-Verlag, Berlín 2006).

Turcotte, D.L. (1986), *Fractals and fragmentation*, J. Geophys. Res. 91, 1921-1926.

Vidal. D., Ridgway, C., Pianet. G., Schoelkopf, J., Roy, R. and Bertrand, F. (2009), *Effect of particle size distribution and packing compression on fluid permeability as predicted by lattice-Boltzmann simulations*, Computers and Chemical Engineering 33, 256-266.

Weibull, W. (1951), *A statistical distribution function of wide applicability*, J. Appl. Mech.-Trans. ASME 18 (3): 293-297.